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NUMERICAL FORMULATION OF COMPOSITION SEGREGATION AT CURVED SOLID-LIQUID INTERFACE DURING STEADY STATE SOLIDIFICATION PROCESS

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INTRODUCTION

The lateral solute segregation that results from a curved solid-liquid interface shape during steady state unidirectional solidification of a binary alloy system has been studied both analytically and numerically by Coriell, Boisvert, Rehm, Sekerka (1). The system under their study is a two dimensional rectangular system. However, most real growth systems are cylindrical systems. Thus, in a previous study(2) we have followed Coriell etc. formalism and obtained analytical results for lateral solute segregation for an azimuthal symmetric cylindrical binary melt system during steady state solidification process. The solid-liquid interface shape is expressed as a series combination of Bessel functions. In this study a computer program has been developed to simulate this lateral solute segregation.

FORMALISM

In this section we present the basic equation and boundary condition used in this calculation. The diffusion equation for an azimuthal system is

$$D\frac{\partial^2 c'(r',z')}{\partial r'^2}+\frac{1}{r'}\frac{\partial c'(r',z')}{\partial r'}+\frac{\partial^2 c'(r',z')}{\partial z'^2}+V\frac{\partial c'(r',z')}{\partial z}=0 \quad \text{---(1)}$$

Where D is diffusivity of solute in the liquid, V is the velocity of solidification,c',r', z',w' are dimensional solute concentration, radial and axial coordinates, and interface thickness. The boundary conditions are

$$VC'_I(k-1)=D\left(\frac{\partial C'(r',z')}{\partial z'_I}-\frac{\partial C'(r',z')}{\partial r'_I}\frac{\partial w'(r')}{\partial r'}\right) \text{-----(2)} \qquad c'(r',z')=c_0 \text{----------(3)}$$

$$\frac{\partial C'(r',z')}{\partial r'_I}=0 \quad \text{at } r'=R \quad \text{--(4)}$$

Where k is distribution coefficient. The variables can be nondimensionalized by letting $c=c'/c_0$, $r=r'/R$, $z=z'/R$, $w=w'/R$ and $\beta=VR/D$ where R is radius of the ampule. The diffusion equation and the boundary conditions become

$$\frac{\partial^2 c(r,z)}{\partial r^2}+\frac{1}{r}\frac{\partial c(r,z)}{\partial r}+\frac{\partial^2 c(r,z)}{\partial z^2}+\beta\frac{\partial c(r,z)}{\partial z}=0 \qquad (5)$$

$$\beta C_I(k-1)=\frac{\partial C(r,z)}{\partial z_I}-\frac{\partial C(r,z)}{\partial r_I}\frac{\partial w(r)}{\partial r} \qquad 6)$$

$$c(r, z=\infty)=1 \qquad (7) \qquad\qquad \frac{\partial C(r,z)}{\partial r}=0 \quad \text{at } r=1 \qquad (8)$$

In the limits of very small β, by using variable separation method, equation (5) can be solved to give

$$c(r,z)=AJ_0(ar)e^{-\frac{b}{2}[1+[1+(2a/b)^2]^{1/2}]z}+A' \qquad (9)$$

From boundary condition $c(r,z=\infty)=1$, We have $A'=1$

From boundary condition $\frac{\partial C(r,z)}{\partial r}=0$ at $r=1$, We have

$$\frac{\partial J_0(ar)}{\partial r}=-aJ_1(ar)=0 \qquad (10)$$

let u_n be the zeros of $J_1(\alpha)$, where n=1 to infinite.

By using p(n) to denote $[1+(\frac{2u_n}{\beta})^2]^{1/2}$. The general solution is

$$C(r,z)=1+\frac{1-k}{k}e^{-\beta(z-w_0)}+\sum_{n=1}^{\infty}A_nJ_0(u_nr)e^{-\frac{\beta}{2}[1+p(n)]^{1/2}z} \qquad (11)$$

There is another boundary condition which needs to be satisfied, i.e.

$$\beta C_I(k-1)=\frac{\partial C(r,z)}{\partial z_I}-\frac{\partial C(r,z)}{\partial r_I}\frac{\partial w(r)}{\partial r'} \qquad (12)$$

The solid-liquid interface shape is assumed to be parabolic δr^2 and is expressed as a series combination of Bessel function, i.e.

$$w(r)=w_0+\sum_{n=1}^{\infty}\delta(n)J_0(u_n r) \qquad \text{------------------------------------(13)}$$

Assume both w(r) and $\frac{dw(r)}{dr}$ to be small, We obtain the solute concentration at the interface to be

$$c_I(r,z)=\frac{1}{k}-\beta\frac{(1-k)}{k}\sum_{n=1}^{\infty}\frac{\delta(n)J_0(u_n r)}{1+2k/[p(n)-1]} \qquad \text{------------------------------(14)}$$

$$c_{SI}(r,z)=kc_I$$

$$=1-\beta(1-k)\sum_{n=1}^{\infty}\frac{\delta(n)J_0(u_n r)}{1+2k/[p(n)-1]} \qquad \text{-----------------------------(15)}$$

This shows that the solute concentration in the solid at the interface is a function of w(r), β and k.

NUMERICAL CALCULATIONS AND RESULTS

The results obtained from these calculations show that

(1) The solute concentration at the interface at r= 0.7 which were calculated by using the general expression eq (11).with n from 1 to i is denoted by $C3_j$. The same value calculated by using the approximate expression eq(15).with n from 1 to i is denoted by $C4_j$ The results in figure 1 show that if we have include more than 40 terms in the calculations, the two expressions give almost exact results.

(2) .The solute concentration in the solid at the interface obtained analytically show that the compositional segregation in the solid is proportional to the deviation of the interface from planarity. The proportional factor being the product of β and (1-k). The solute concentration at the interface $C_{si}(x)$ calculated by using the general expression and by using the approximate expression for a small β limit agree very well.The results for β =0.173, k=4 and δ = 0.05, 0.1, 0.2, 0.375 and 0.4 are calculated and the result for δ =0.375 is shown in figure 2.The results of $C1_j$ were calculated by using the general expression eq (11) with n from 1 to 89, while the results of $C2_j$ were calculated by using the approximate

expression eq(15) with n from 1 to 89.

3. The results in figure 3 show that the solute concentration at the interface $C_{si}(x)$ is linearly proportional to the amplitude of the interface shape deviation at both x=0, the center of the interface and x=1, the edge of the interface for β =0.1256637, 1.256639 and 12.56637. Theseresults are calculated by using the approximate expression eq(15)which is valid for small β. The results for β =12.56637 need to be reconsidered.
4. The results in figure 4 show that the solute concentration at the interface $C_{si}(x)$ for different interface amplitudes all have the same shape as a function of its radius.
5. In table 1, for various values of k, β, and δ, we give the solute concentration at the interface $C_{si}(x)$ in the solid at x=o and x=1. These results have similar β and k dependent as that in table 1 in ref 1.

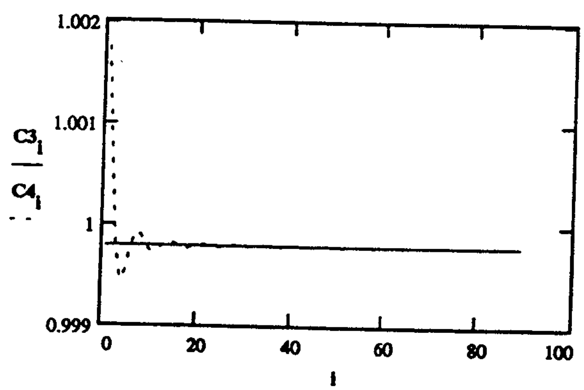

Figure 1. Caption see text.

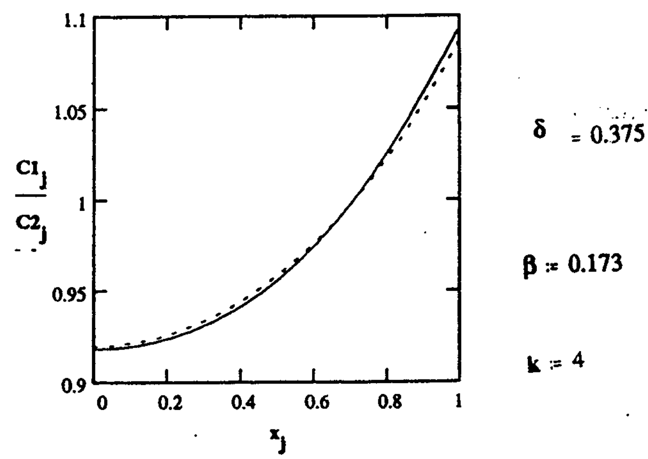

Figure 2. Caption see text.

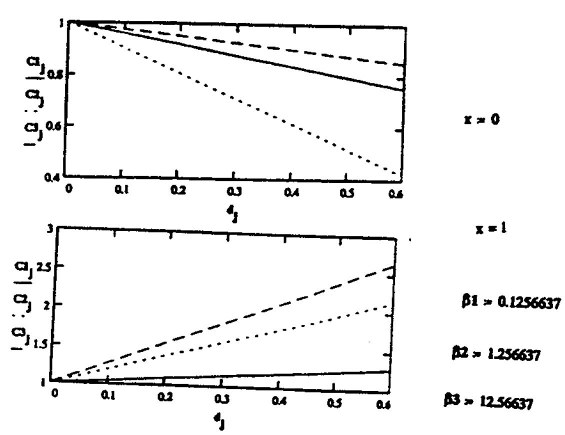

Figure 3. Caption see text.

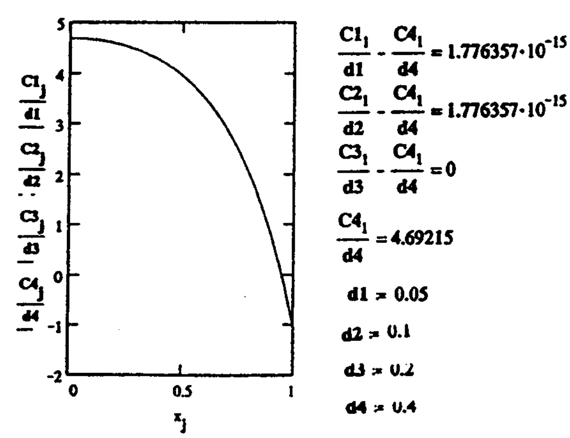

Figure 4 Caption see text.

k	$\frac{\beta}{4\cdot 3.14}$	δ	$C_{si(0)}$	$C_{si(1)}$
0.1	0.01	0.05	0.9972	1.003
0.1	0.01	0.1	0.9944	1.006
0.1	0.01	0.2	0.9887	1.011
0.1	0.01	0.3	0.9831	1.017
0.1	0.01	0.4	0.9775	1.022
0.1	0.01	0.5	0.9719	1.028
0.1	0.01	0.6	0.9662	1.034
0.1	0.01	0.7	0.9606	1.039
0.1	0.01	0.8	0.95498	1.045
0.1	0.01	0.9	0.9494	1.05
0.1	0.01	1.0	0.9437	1.056

k	$\frac{\beta}{4\cdot 3.14}$	δ	$C_{si(0)}$	$C_{si(1)}$
2	0.01	0.05	1.0029	0.997
2	0.01	0.1	1.0058	[illegible]
2	0.01	0.2	1.0117	0.988
2	0.01	0.3	1.0175	0.982
2	0.01	0.4	1.0233	0.976
2	0.01	0.5	1.0292	[illegible]
2	0.01	0.6	1.0350	0.964
2	0.01	0.7	1.0408	0.958
2	0.01	0.8	1.0467	0.952
2	0.01	0.9	[illegible]	0.946
2	0.01	1.0	1.0583	0.94

k	$\frac{\beta}{4\cdot 3.14}$	δ	C	$C_{si(1)}$
2	0.1	0.05	[illegible]	0.979
2	0.1	0.1	1.0326	0.958
2	0.1	0.2	1.0652	0.916
2	0.1	0.3	[illegible]	0.873
2	0.1	0.4	[illegible]	0.831
2	0.1	0.5	1.163	0.789
2	0.1	0.6	1.1957	0.747
2	0.1	0.7	1.2283	0.705
2	0.1	0.8	1.2609	0.663
2	0.1	0.9	1.2935	0.62
2	0.1	1.0	1.3261	0.578

k	$\frac{\beta}{4\cdot 3.14}$	δ	$C_{si(0)}$	$C_{si(1)}$
0.1	0.1	0.05	0.973	1.027
0.1	0.1	0.1	0.9459	1.055
0.1	0.1	0.2	0.8918	1.109
0.1	0.1	0.3	0.8378	1.164
0.1	0.1	0.4	0.7837	1.218
0.1	0.1	0.5	0.7296	1.273
0.1	0.1	0.6	0.67553	1.328
0.1	0.1	0.7	0.62145	1.382
0.1	0.1	0.8	0.5674	1.437
0.1	0.1	0.9	0.5133	1.491
0.1	0.1	1.0	0.4592	1.546

k	$\frac{\beta}{4\cdot 3.14}$	δ	$C_{si(0)}$	$C_{si(1)}$
10	0.01	0.05	1.0204	0.977
10	0.01	0.1	1.0408	0.954
10	0.01	0.2	1.0815	0.908
10	0.01	0.3	1.1223	0.862
10	0.01	0.4	1.163	0.817
10	0.01	0.5	1.2038	0.771
10	0.01	0.6	1.2445	0.725
10	0.01	0.7	1.2853	0.679
10	0.01	0.8	1.3260	0.633
10	0.01	0.9	1.3668	0.587
10	0.01	1.0	1.4076	0.542

k	$\frac{\beta}{4\cdot 3.14}$	δ	$C_{si(0)}$	$C_{si(1)}$
10	0.1	0.05	1.0481	0.905
10	0.1	0.1	1.09627	0.809
10	0.1	0.2	1.1925	0.619
10	0.1	0.3	1.2888	0.428
10	0.1	0.4	1.385	0.237
10	0.1	0.5	1.4814	0.047
10	0.1	0.6	1.5776	-0.144
1.0	0.1	0.7	1.6739	-0.335
10	0.1	0.8	1.7701	-0.525
10	0.1	0.9	1.8664	-0.716
10	0.1	1.0	1.9627	-0.907

Table 1. Solute segregation for solid-liquid interface shape $w(r)= \delta r^2$ for various values of k, β, and δ

REFERENCES

1. Coriell, S. R., F. Boisvert, R. G. Rehm, and R. F. Sekerka, "Lateral Solute Segregation During Unidirectional Solidification of a Binary Alloy with a Curved Solid-Liquid interface", Journal of Crystal Growth 54, 167, 1981

2. Wang, J. C., "Investigation of Compositional Segregation During Unidirectional Solidification of Solid Solution Semi-Conducting Alloy", NASA CR 162051, ed. by Karr, G. R., Barfield, J., and Kent, M., pXLI 1-20, Aug. 1982.